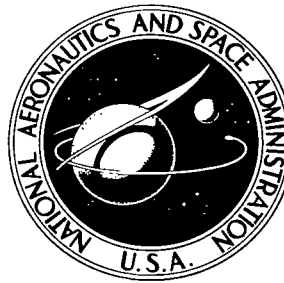


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THERMODYNAMIC PROPERTIES OF LIQUID METAL SOLUTIONS IN THE CESIUM-MERCURY SYSTEM

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THERMODYNAMIC PROPERTIES OF LIQUID METAL SOLUTIONS IN THE CESIUM-MERCURY SYSTEM

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SUMMARY

Measurements were made of the electromotive force of cesium-cesium amalgam cells, over the composition range from 0.05 to 0.15 atomic fraction of cesium, and at temperatures of 215°, 240°, and 265° C (488, 513, and 538 K). The thermodynamic activity of cesium in the amalgam a_{Cs} was calculated from the relation $nF\epsilon = -RT \ln a_{\text{Cs}}$, where n is the number of electrons in the stoichiometric reaction, F is the faraday, 23 066 calories per volt ($9.6487 \times 10^4 \text{ C mol}^{-1}$), ϵ is the electromotive force in volts, R is the gas constant in calories per mole degree ($8.3143 \text{ (J)(K}^{-1}\text{)(mol}^{-1}\text{)}$) and T is the temperature in K.

Partial Gibbs free energies of cesium $\Delta\bar{G}_{\text{Cs}}$, partial entropies of cesium $\Delta\bar{S}_{\text{Cs}}$, and partial heats of cesium $\Delta\bar{H}_{\text{Cs}}$ of mixing in cesium amalgam were calculated. The partial excess free energy of cesium $\Delta\bar{G}_{\text{Cs}}^e$ and partial excess entropy of cesium $\Delta\bar{S}_{\text{Cs}}^e$ were also calculated.

The activity of the other component, mercury, was obtained by means of the Gibbs-Duhem relation. From this, the partial free energy of mixing of mercury in cesium was obtained. Partial entropies and heats of mixing were calculated. Integral values of the heats, entropies, and free energies were obtained by graphical integration or by calculation from the partial quantities. Over the composition range studied, the activity of cesium exhibited large negative deviations from ideality, while the activity of mercury, above 0.93 atomic fraction, deviated less than 10 percent from Raoult's law. The enthalpies and free energies of mixing were large and negative, even in dilute cesium-mercury solutions. It is concluded that the deviations from the ideal solution are probably due to specific compound formation and the results can be explained quantitatively on this basis.

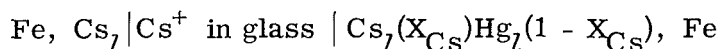
INTRODUCTION

There is little thermodynamic data for liquid cesium-mercury solutions. Bent and

Hildebrand (ref. 1) measured the vapor pressures of some cesium amalgams over the composition range from 0.06 to 0.29 atomic fraction of cesium and at temperatures from 281° to 378° C (554 to 651 K). The partial thermodynamic quantities of mercury based on these measurements have been derived by Kubaschewski and Catterall (ref. 2) and by Hultgren et al. (ref. 3). The partial values are those for the process in which one component is dissolved in an infinite amount of alloy. The integral values are those that are involved in the formation of an alloy from its constituent elements. The partial excess free energy $\Delta\bar{G}_{Cs}^e$ and partial heat of mixing $\Delta\bar{H}_{Cs}$ of cesium at infinite dilution were obtained from the electromotive force (emf) of a cesium-cesium amalgam cell over the range 25° to 35° C (298 to 308 K) at a single amalgam composition of 0.0028 atomic fraction of cesium (ref. 4). The partial heat of mixing of cesium was also obtained calorimetrically (ref. 5) at 25° C (298 K) and infinite dilution. There was fair agreement with the emf data. Better agreement was obtained using an amalgam partition method together with tabulated emf data (ref. 6). The value for $\Delta\bar{G}_{Cs}^e$ at infinite dilution obtained in this way was in good agreement with the value reported by Bent, Forbes, and Forziati (ref. 4).

In view of the absence of reported emf data on dilute amalgams at other than room temperature, the present investigation was carried out. The emf of cesium-cesium amalgam cells was measured over the composition range from 0.05 to 0.15 atomic fraction of cesium X_{Cs} and at temperatures of 215°, 240°, and 265° C (488, 513, and 538 K), all chosen to be above the melting point of the highest melting alloy of cesium and mercury ($CsHg_2$ melting point, 208° C (or 481 K)).

The thermodynamic properties of the liquid alloys were calculated from the emf values and their temperature coefficients for concentration cells of the type



in which the change of state in the cell may be expressed by the equation $Cs_l = Cs$ (in the liquid amalgam) or $Cs_l(X_{Cs})Hg_l(1 - X_{Cs})$. The electrode reactions are $Cs_l \rightarrow Cs^+ + e$ and $Cs^+ + e \rightarrow Cs_l(X_{Cs})Hg_l(1 - X_{Cs})$. The state of each pure component at the given temperature was taken as the standard state. The activity of cesium, a_{Cs} , was calculated from the relation

$$nF\epsilon = -RT \ln a_{Cs}$$

or

$$\log a_{Cs} = \frac{-5040\epsilon}{T}$$

where ϵ is the electromotive force (V) and T is the absolute temperature (K).

The partial values of free energy $\Delta\bar{G}_{\text{Cs}}$, entropy $\Delta\bar{S}_{\text{Cs}}$, and heat of mixing $\Delta\bar{H}_{\text{Cs}}$, for cesium were calculated from the following equations:

U. S. Customary	SI Units
$\Delta\bar{G}_{\text{Cs}} = -23\,066 \epsilon \text{ cal/g-atom}$	$\Delta\bar{G}_{\text{Cs}} = -9.6487 \times 10^4 \epsilon \text{ J/g-atom}$
$\Delta\bar{S}_{\text{Cs}} = 23\,066 \frac{d\epsilon}{dT} \text{ cal/(g-atom)(deg)}$	$\Delta\bar{S}_{\text{Cs}} = 9.6487 \times 10^4 \frac{d\epsilon}{dT} \text{ J/(g-atom)(deg)}$
$\Delta\bar{H}_{\text{Cs}} = 23\,066 \left(T \frac{d\epsilon}{dT} - \epsilon \right) \text{ cal/g-atom}$	$\Delta\bar{H}_{\text{Cs}} = 9.6487 \times 10^4 \left(T \frac{d\epsilon}{dT} - \epsilon \right) \text{ J/g-atom}$

The integral values for the heat of mixing ΔH were obtained by graphical integration of the equation (ref. 7)

$$\Delta H = X_{\text{Hg}} \int_0^x \Delta\bar{H}_{\text{Cs}} dx$$

where

$$x = \frac{X_{\text{Cs}}}{X_{\text{Hg}}}$$

The integral values for the free energy of mixing were obtained from the equation

$$\Delta G = X_{\text{Cs}} \Delta\bar{G}_{\text{Cs}} + X_{\text{Hg}} \Delta\bar{G}_{\text{Hg}}$$

where ΔG is the integral value and $\Delta\bar{G}_{\text{Cs}}$ and $\Delta\bar{G}_{\text{Hg}}$ are the partial quantities. The integral values for the entropy of mixing were obtained from a similar equation.

To calculate the activity of mercury, the activity coefficient of mercury was obtained by graphical integration of the Gibbs-Duhem equation

$$\log \gamma_{\text{Hg}} = - \int_0^{X_{\text{Cs}}} \frac{X_{\text{Cs}}}{X_{\text{Hg}}} d \log \gamma_{\text{Cs}}$$

where γ_{Hg} is the activity coefficient, equal to the ratio of activity to atomic fraction. The partial free energy of mercury was obtained from the activity, the partial heat from the integral heat, and the partial entropy from the partial free energy and partial heat.

Values of the excess thermodynamic quantities were calculated. The excess function is equal to the difference between the total and ideal value of the function. Thus, in general,

$$\Delta Z = \Delta Z^i + \Delta Z^e$$

where ΔZ is the value of the thermodynamic function for the solution, ΔZ^i is the value that the function ΔZ would have if the solution were ideal, and ΔZ^e is the difference between the two, the excess quantity.

The partial excess free energy of cesium was obtained from the equation

$$\begin{aligned} \Delta \bar{G}_{\text{Cs}}^e &= \Delta \bar{G}_{\text{Cs}} - RT \ln X_{\text{Cs}} \\ &= RT \ln \gamma_{\text{Cs}} \text{ cal/g-atom (or J/(g-atom))} \end{aligned}$$

and the partial excess free energy of mercury was obtained from a similar equation. The partial excess entropy of cesium was obtained from

$$\Delta \bar{S}_{\text{Cs}}^e = \Delta \bar{S}_{\text{Cs}} + R \ln X_{\text{Cs}} \text{ cal/(g-atom)(deg) (or J/g-atom)}$$

and the partial excess of mercury from a similar equation. The integral excess values were obtained from the partial excess values.

EXPERIMENTAL PROCEDURE

The design of the measurement cell and the methods used were essentially the same as described in earlier investigations (refs. 8 and 9). The cell consisted of two borosilicate glass tubes, each sealed at the bottom, and one placed inside the other. The inner tube was 300 millimeters long and 11 millimeters in diameter; the outer tube was 70 millimeters long and 22 millimeters in diameter. The inner tube, which served as the electrolyte, was reduced to a thickness of about 1/2 millimeter at the closed end. In thinning and closing this tube, a small amount of cesium oxide was incorporated into the seal to provide conduction by cesium ions. Pure cesium was placed in the inner tube and a cesium alloy of known composition in the outer tube. These two tubes were then placed inside a third tube (280 mm long and 28 mm in diam) which was equipped with a

side arm for blanketing the cell with pure argon. Pure iron wires passing through rubber stoppers served as leads. The cell was placed in an electric furnace consisting of a copper tube wound with Nichrome wire. The temperature was controlled to within $\pm 1^{\circ}$ C by a temperature indicator controller and was measured with an iron-constantan thermocouple connected to a potentiometer. The potentiometer, with an electrometer as an impedance-matching amplifier, was also used to measure the electromotive-force of the amalgam cell to within ± 1 millivolt.

The experiments were conducted in a dry box in an atmosphere of purified argon. Cesium of at least 99.9 percent purity was used; the mercury was triple distilled. The alloys were prepared in the outer tube by mixing weighed amounts of cesium and mercury. For each sample, an emf reading was obtained at the three temperatures, with the temperature held constant for at least 20 minutes before the reading was taken. The potential did not vary more than ± 2 millivolts during this interval. The electromotive-force values used in the calculations were average values taken from measurements on three separate cells.

A consideration of the uncertainty in the activity of the amalgam resulting from the experimental uncertainties in the temperature measurement ($\pm 1^{\circ}$ C) and the emf measurement (± 1 mV) was undertaken through a propagation of errors treatment. The results showed that in every case these uncertainties were negligible in comparison with the influence of the ± 2.5 percent variation in electromotive force noted from cell to cell at each composition and temperature.

RESULTS AND DISCUSSION

Values of the electromotive force of the cesium-cesium amalgam cell are given in table I. The values are the mean and the standard deviation of the mean. Included in the table are the temperature coefficients of the electromotive force. These were obtained by fitting a straight line to the experimental points by the least squares method. The values shown are the slope and the standard deviation of the slope.

In figure 1 activities are plotted as a function of the atomic fraction of cesium. The dashed lines in figures 1(b) and (c) show the activities expected if the solutions were ideal and obeyed Raoult's law. Figures 1(a) and (b) show the calculated activities of cesium and mercury, respectively, at 240° C (513 K), obtained from this study. The activities are, in all instances, less than they would be if Raoult's law were obeyed. The activity of cesium deviates from the ideal by a factor as large as 10^6 . However, in the region rich in mercury (i. e., above an atomic fraction of 0.93 of mercury), the departure of the activity of mercury from the law becomes small, deviating less than 10 percent.

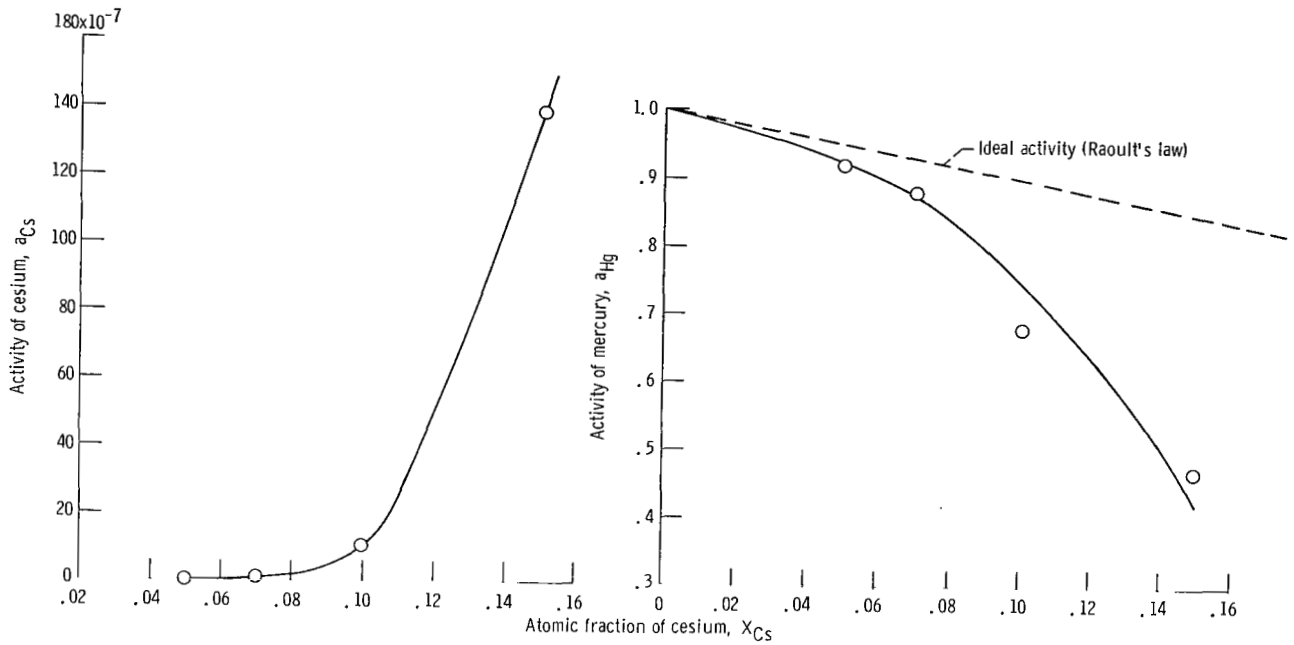
TABLE I. - VALUES AND TEMPERATURE COEFFICIENTS
OF ELECTROMOTIVE FORCE

Atomic fraction of cesium, X_{Cs}	Temperature, °C (K)			Temperature coefficient of electromotive force, $d\epsilon/dT$, V/deg
	215 (488)	240 (513)	265 (538)	
	Electromotive force, V			
0.05	0.759±0.003	0.754±0.004	0.751±0.006	-1.6±0.7×10 ⁻⁴
.07	.740±0.016	.727±0.014	.719±0.013	-4.2±2.2×10 ⁻⁴
.10	.620±0.009	.607±0.006	.595±0.014	-5.0±1.3×10 ⁻⁴
.15	.515±0.009	.500±0.009	.488±0.008	-5.4±1.4×10 ⁻⁴

The calculated thermodynamic quantities for the liquid cesium-mercury alloys at 240° C (513 K) are summarized in table II. The values are mean values with the standard deviation of the mean. The thermodynamic data are shown in figure 2. The partial and integral free energies of mixing at 240° C (513 K) are plotted in figure 2(a); the excess free energy values are shown in figure 2(b). The partial and integral molar entropies of mixing are plotted in figure 2(c); the excess entropy values are shown in figure 2(d). Figure 2(e) shows the partial and integral heats of mixing.

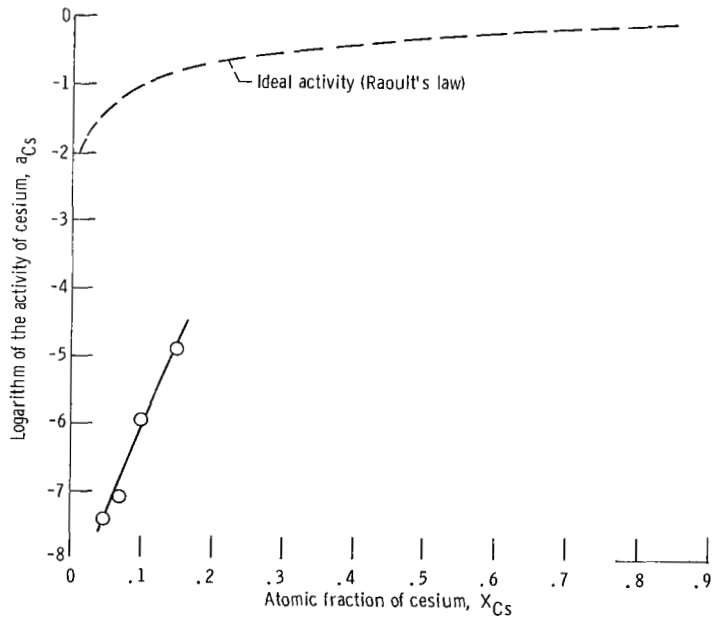
The behavior of alkali metal - mercury alloys may be attributed to the existence of structural groups of metallic compounds in the liquid phase. The phase diagram shows that at least seven intermediate phases are formed (ref. 3). Bent and Hildebrand (ref. 1) showed that approximate values for the activity of mercury in sodium-mercury alloys can be obtained by assuming the formation of any one compound of sodium and mercury. It was observed that over a limited range of composition the calculated activity function coincided closely with the smooth curve through the experimental points. Some deviations could be accounted for by assuming dissociation of the compound. A more general calculation was based on the assumption of several definite compounds, each dissociating in accord with the law of mass action, and each molecular species obeying Raoult's law. Thus, for a system composed of 1 mole of sodium and n moles of mercury (ref. 1),

$$n = \frac{a}{1-a} + \frac{2K_2a^2 + 4K_4a^4 + 6K_6a^6}{1 + K_2a^2 + K_4a^4 + K_6a^6}$$



(a) Cesium.

(b) Mercury (experimental values compared with ideal activity).



(c) Cesium (experimental values compared with ideal activity).

Figure 1. - Activities of cesium and mercury at 240° C (513 K).

TABLE II. - THERMODYNAMIC DATA FOR LIQUID CESIUM-MERCURY ALLOYS AT 240° C (513 K)

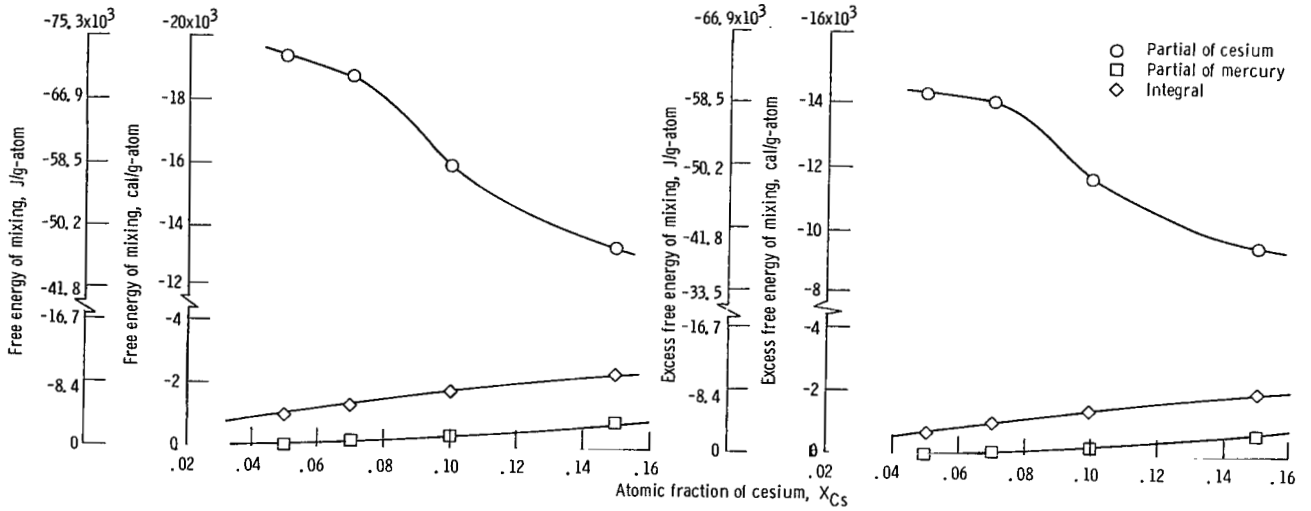
(a) U. S. Customary units

Thermo-dynamic properties	Atomic fraction of cesium, X_{Cs}					
	0.05	0.07	0.10	0.15	0.2	0.3
$\Delta\bar{G}_{Cs}$	-17 390±90	-16 770±320	-14 000±140	-11 530±200	-----	-----
$\Delta\bar{G}_{Hg}$	-85±80	-125±100	-400±100	-760±130	-----	-----
ΔG	-950±80	-1 290±115	-1 760±105	-2 380±140	-----	-----
$\Delta\bar{G}_{Cs}^e$	-14 300±90	-14 100±320	-11 650±140	-9 550±200	-----	-----
$\Delta\bar{G}_{Hg}^e$	-35±30	-55±50	-290±75	-600±120	-----	-----
ΔG^e	-750±30	-1 040±70	-1 430±80	-1 950±130	-----	-----
$\Delta\bar{S}_{Cs}$	-3.7±1.7	-9.7±5.1	-11.4±3.0	-12.5±3.2	-----	-----
$\Delta\bar{S}_{Hg}$	-0.11±0.01	0.00±0.05	-0.88±0.08	-1.85±0.18	-----	-----
ΔS	-0.30±0.03	-0.67±0.06	-1.64±0.16	-3.45±0.34	-----	-----
$\Delta\bar{S}_{Cs}^e$	-9.7±1.7	-15.0±5.1	-16.0±3.0	-16.3±3.2	-----	-----
$\Delta\bar{S}_{Hg}^e$	-0.21±0.02	-0.14±0.05	-1.09±0.10	-2.17±0.20	-----	-----
ΔS^e	-0.70±0.07	-1.17±0.11	-2.29±0.22	-4.29±0.42	-----	-----
$\Delta\bar{H}_{Cs}$	-19 280±870	-21 730±2620	-19 860±1530	-17 920±1640	-----	-----
$\Delta\bar{H}_{Hg}$	25±2	175±25	-15±3	-285±25	-----	-----
ΔH	-940±45	-1 360±180	-2 000±150	-2 930±250	-----	-----
γ_{Cs}	0.78±0.08×10 ⁻⁶	0.10±0.03×10 ⁻⁵	0.11±0.02×10 ⁻⁴	0.80±0.16×10 ⁻⁴	-----	-----
γ_{Hg}	0.97±0.09	0.95±0.09	0.75±0.07	0.56±0.06	-----	-----
a_{Cs}	0.39±0.04×10 ⁻⁷	0.72±0.23×10 ⁻⁷	0.11±0.02×10 ⁻⁵	0.12±0.02×10 ⁻⁴	-----	-----
a_{Hg}	0.92±0.09	0.88±0.09	0.68±0.07	0.47±0.05	-----	-----
			^a 0.675		^a 0.241	^a 0.058±0.005

(b) SI Units

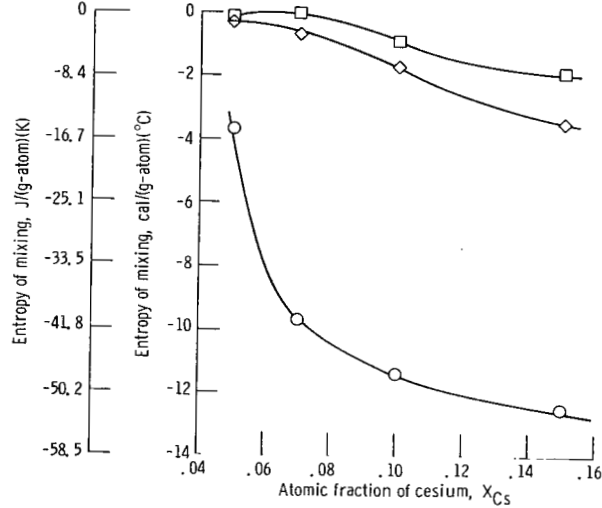
	0.05	0.07	0.10	0.15
$\Delta\bar{G}_{Cs}$	-72 760±380	-70 170±1 340	-58 580±590	-48 240±840
$\Delta\bar{G}_{Hg}$	-355±335	-525±420	-1 675±420	-3 180±540
ΔG	-3 980±335	-5 400±480	-7 360±440	-9 960±590
$\Delta\bar{G}_{Cs}^e$	-59 830±380	-58 990±1 340	-48 740±590	-39 960±840
$\Delta\bar{G}_{Hg}^e$	-145±125	-230±210	-1 210±310	-2 510±500
ΔG^e	-3 140±125	-4 350±290	-5 980±335	-8 160±540
$\Delta\bar{S}_{Cs}$	-15±7	-41±21	-48±13	-52±13
$\Delta\bar{S}_{Hg}$	-0.46±0.04	0.00±0.21	-3.7±0.3	-7.7±0.8
ΔS	-1.3±0.1	-2.8±0.3	-6.9±0.7	-14±1
$\Delta\bar{S}_{Cs}^e$	-41±7	-63±21	-67±13	-68±13
$\Delta\bar{S}_{Hg}^e$	-0.88±0.08	-0.59±0.21	-4.6±0.4	-9.1±0.8
ΔS^e	-2.9±0.3	-4.9±0.5	-9.6±0.9	-18±2
$\Delta\bar{H}_{Cs}$	-80 670±3640	-90 920±10 960	-83 090±6400	-74 980±6860
$\Delta\bar{H}_{Hg}$	105±8	730±105	-63±13	-1 190±105
ΔH	-3 930±190	-5 690±750	-8 370±630	-12 260±1050

^aRef. 3. Data taken at 327° C (600 K).



(a) Free energy of mixing.

(b) Excess free energy of mixing.



(c) Entropy of mixing.

Figure 2 - Thermodynamic data.

where a is the activity of mercury in the solution, and

$$K_2 = \frac{a_{NaHg_2}}{a_{Na}^2 a_{Hg}} \quad K_4 = \frac{a_{NaHg_4}}{a_{Na}^4 a_{Hg}} \quad K_6 = \frac{a_{NaHg_6}}{a_{Na}^6 a_{Hg}}$$

With this approach good agreement was obtained over the whole composition range of interest.

A similar treatment can be applied to cesium-mercury alloys. For the formation

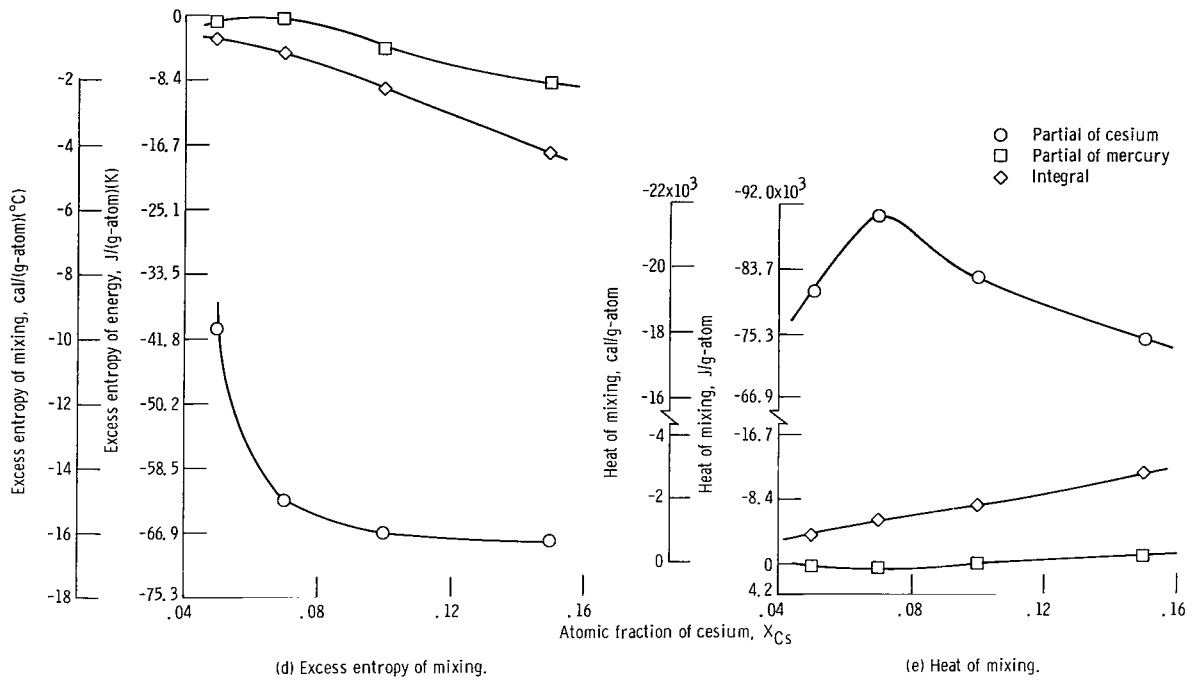


Figure 2. - Concluded.

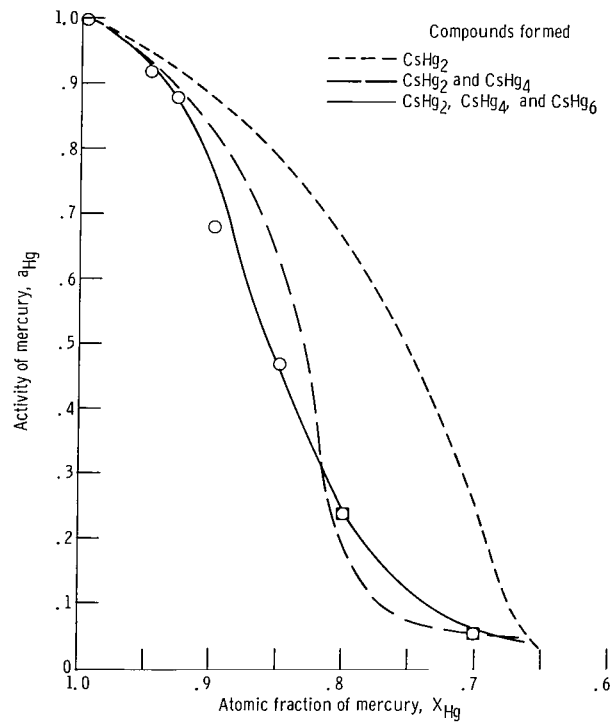


Figure 3. - Activity of mercury, calculated by assuming compound formation, compared with experimental values.

of CsHg_2 alone, the activity of mercury was calculated and the results are shown in figure 3. The results obtained by assuming the formation of CsHg_2 and CsHg_4 are also shown in figure 3. The fits to the experimental data are not satisfactory. It was necessary, therefore, to consider three compounds of cesium and mercury, CsHg_n ($n = 2, 4, 6$), in equilibrium with the components of the liquid-metal solution in order to obtain an acceptable fit to the experimental results. The formation of these compounds has been assumed because the Liquidus curve for cesium-mercury alloys shows three maxima corresponding to these compounds (ref. 10). Thus, a smooth curve through the experimental points is calculated by fitting values to all three equilibrium constants. The experimental data used for the curve fitting included not only those from this study but also the results of Hultgren et al. (ref. 3) for the activity of mercury at 327°C (600 K). These could be included since the change in activity with temperature is very small (ref. 1). These values are listed in table II. The values for the equilibrium constants $K_2 = 2.6 \pm 0.2 \times 10^3$, $K_4 = 1.3 \pm 0.2 \times 10^5$, and $K_6 = 3.8 \pm 0.6 \times 10^5$ were obtained from the equation using a method of least squares. The deviation for the solid curve shown in figure 3 is 0.0137.

CONCLUSIONS

The results of this investigation show that, even in dilute cesium-mercury solutions, very substantial deviations from the laws of ideal dilute solutions occur, in that the partial heats and partial free energies of mixing of cesium are large and negative. These large negative deviations from ideality can be explained quantitatively on the basis of specific compound formation. Three compounds CsHg_n ($n = 2, 4, 6$) had to be assumed to adequately explain the experimental results. Their respective formation constants are $2.6 \pm 0.2 \times 10^3$, $1.3 \pm 0.2 \times 10^5$, and $3.8 \pm 0.6 \times 10^5$ at 240°C (513 K).

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, September 17, 1969,
120-34.

APPENDIX - SYMBOLS

<p>a_j activity of component j</p> <p>F Faraday constant, 23 066 cal/V; 9.6487×10⁴ C-mol⁻¹</p> <p>ΔG integral free energy of mixing</p> <p>$\Delta \bar{G}$ partial free energy of mixing</p> <p>ΔH heat of mixing</p> <p>$\Delta \bar{H}$ partial heat of mixing</p> <p>n number of electrons in stoichiometric reaction</p> <p>R gas constant, cal/deg, J/deg</p> <p>ΔS integral entropy of mixing</p> <p>$\Delta \bar{S}$ partial entropy of mixing</p> <p>T temperature, K</p>	<p>X_j atomic fraction of component j</p> <p>x ratio of atomic fractions</p> <p>γ_j activity coefficient of component j</p> <p>ϵ electromotive-force, V</p> <p>ΔZ integral function of state</p> <p>Subscripts:</p> <p>Cs cesium</p> <p>Hg mercury</p> <p>l liquid</p> <p>Superscripts:</p> <p>e excess</p> <p>i ideal</p>
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